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MIHAN et al.

Serial No. 09/937,844

Filed: 9/28/2001

For: POLYMERIZATION OF OLEFINS

Honorable Commissioner of

12/15/2003 DTESSEM1 00000082 09937844

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Art Unit: 1713

Examiner: Lu, C.C.

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BRIEF ON APPEAL

Sir:

This appeal is from the examiner's final rejection of claims 1-12 dated June 18, 2003.

REAL PARTY IN INTEREST

The real party in interest is BASF Aktiengesellschaft of Ludwigshafen, Germany.

Reel 012313, Frame 0190, recorded on 9/28/2001.

RELATED APPEALS AND INTERFERENCES

To appellants' knowledge and belief, there are no interferences or other appeals which will directly affect or be directly affected by or have a bearing on the Board's decision in this application.

STATUS OF CLAIMS

Claims 1-12 remain in the application and stand rejected under 35 USC §103(a) as being unpatentable over Tani et al. (JP 10-231317).

STATUS OF AMENDMENTS

An amendment after final was filed on October 17, 2003 in which new claims 14 and 15 were introduced. In the advisory action dated November 18, 2003, the examiner did not indicate that these claims have *not* been entered. Applicants assume, therefore, that they have been entered. These claims have not been rejected.

SUMMARY OF INVENTION

The present claims are drawn to processes for polymerization of olefins in which the polymerization is carried out in the presence of catalysts having at least one complex composed of a transition metal and a tridentate macrocyclic ligand bearing at least one substituent having a donor function. The catalyst may include one or more activator compounds. Additionally, the present claims specify certain ligand and transition metal components of the catalyst complex, certain activator compounds and olefins, and certain further reaction conditions. Finally, the claims are also drawn to a catalyst system having at least one transition metal complex as described above and at

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least one activator compound.

ISSUES

Whether claims 1-12 are unpatentable under 35 USC § 103 as being obvious from the disclosure of Tani et al. (JP 10-231317).

GROUPING OF CLAIMS

Other than a specific reference to claim 11, the examiner's arguments do not differentiate in their application between any of claims 1-12. Applicants' replies to date have been directed to the examiner's general arguments, and are so directed herein.

ARGUMENTS

The following legal authorities are relied on in the following arguments:

In re Oetiker, 977 F.2d 1443, 24 USPQ2d 1443 (Fed. Cir. 1992)

In re Chupp, 816 F.2d 643, 2 USPQ2d 1437 (Fed. Cir. 1987)

THE REJECTIONS

As indicated above, the examiner has rejected claims 1-12 under 35 USC §103(a) as obvious over the disclosure of Tani, et al. (JP 10-231317). The applicants have traversed this rejection on the basis of objective evidence showing unexpected results. Such objective evidence must be considered in the ultimate determination of

patentability, and unexpected superiority can rebut a *prima facie* case of obviousness (*In re Oetiker*, 977 F.2d 1443, 24 USPQ2d 1443 (Fed. Cir. 1992), *In re Chupp*, 816 F.2d 643, 2 USPQ2d 1437 (Fed. Cir. 1987)). As applicants have indicated in their previous replies, the presently claimed invention produces polymers which have a greatly elevated molecular weight when compared with those of the prior art. These results are unexpected given the structural similarity of the catalyst complexes.

Examples 1-8 of Tani utilize catalysts comparable to those of the present claims. Examples 1-4 and 7-8 employ catalysts A-C and E, which are 1,3,5-triisopropylhexahydro-1,3,5-triazines. Example 5-6 employ catalyst D, which is a 1,3,5-trimethylhexahydro-1,3,5-triazine. The table on page 29 of Tani indicates comparable polymerization results. In Ex.3-6 and 8 the polymerization reaction was carried out using catalysts B-E, with a reaction time of one hour. These processes gave reaction products having high levels of C₄₋₁₄ oligomers. In ex.7, the reaction time was increased to three hours and catalysis was performed using catalyst E. The reaction product from this process was polymer of an unspecified melting point or carbon number. Examples 1-2 employed catalyst A, and in reaction times of four and two hours, respectively, resulted in small amounts of polymer, again of unspecified melting point or carbon number. Thus, Tani produces either C₄₋₁₄ oligomer fractions predominantly or significantly, or with longer residence times, produces some polymer.

The present specification's example 8 describes a 1,3-dimethyl-5-(2-hydroxyethyl)-1,3,5-triazacyclohexane for use in a catalyst, which differs only slightly

from the 1,3,5-trimethylhexahydro-1,3,5-triazine of catalyst D in examples 5-6 of Tani. Present examples 20 and 21 are comparable processes to Tani's examples 5-6, and yet the differences are striking. Rather than significant oligomer fractions, after a one hour residence time the present process produced polymers having molecular weights of 46,729 and 36,659 g/mol, respectively. Such high molecular weight polymers obtained in such a relatively short residence time would not have been expected from catalysts of such a similar nature to those of Tani.

Present examples 9-10 employ catalysts having 1-(2-oxidoctyl)-1,4,7-triazacyclononane, and the present specification reports comparative data from polymerization utilizing catalysts which differ in the same manner as those compared above. The comparative N,N',N''-trimethyl-1,4,7-triazacyclononane ligand-containing catalysts yield only dimers (p.13:24-31). The results of present examples 9-10, in contrast, yield polymers having molecular weights of 205,282 and 139,795 g/mol, respectively. Such vast increases in molecular weight are entirely unexpected.

Additionally, the processes in examples 11-13, 16-17, and 19 each produced polymers having melting points which demonstrate that no significant amounts of oligomers were produced. The relevant melting points range from 126.1°C to 138.0°C, and with C₆₀ polymers melting on average at nearly 100°C, the average molecular carbon count in the resulting polymers is quite high. Accordingly, the demonstrated unexpected results exist across the entire scope of the present claims.

The examiner's reply to these comparative results in the advisory action of

November 18, 2003 is that the present working examples “do not give good polymer productivity.” However, it is not clear that the data allow such a comparison, and in any event, this is not the point. The point of the applicants’ presentation is that for producing high molecular weight polymers, the present processes and catalysts are vastly superior, and unexpectedly so, to those of the sort disclosed in Tani. The catalysts according to the prior art produce dimers or oligomers under comparable reaction times, where the present catalysts produce high molecular weight polymers. This is the difference, and it is substantial and unexpected. Accordingly, the examiner’s *prima facie* showing of obviousness is rebutted. No substantive reply has been set forward to challenge the present showing.

CONCLUSION

As the present comparative data demonstrate significantly improved and unexpected results in molecular weight of the relevant polymers, the examiner’s *prima facie* case of obviousness is rebutted and should be withdrawn. Applicants respectfully request that the rejection be withdrawn and that the case be passed to the examiner for allowance.

Please charge any shortage in fees due in connection with the filing of this paper, including Extension of Time fees to Deposit Account No. 11-0345. Please credit any excess fees to such deposit account.

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Respectfully submitted,

KEIL & WEINKAUF

A handwritten signature in black ink, appearing to read 'David C. Liechty', with a stylized, cursive script.

David C. Liechty
Reg. No. 48,692

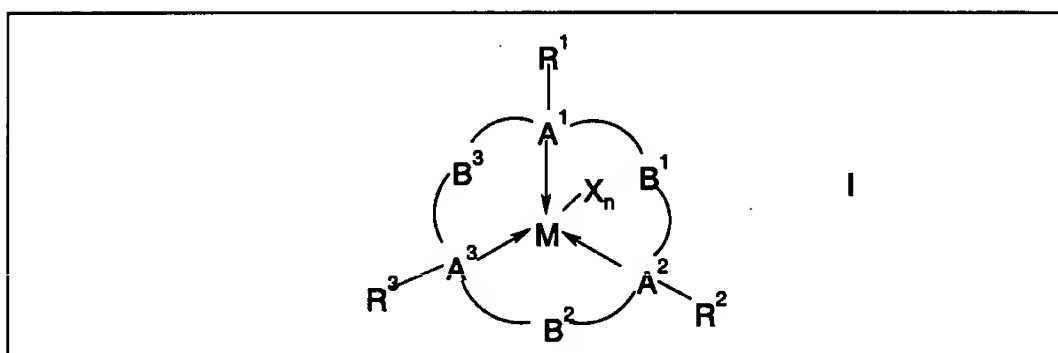
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APPENDIX

Claims in Mihan et al. Ser. No. 09/937,844

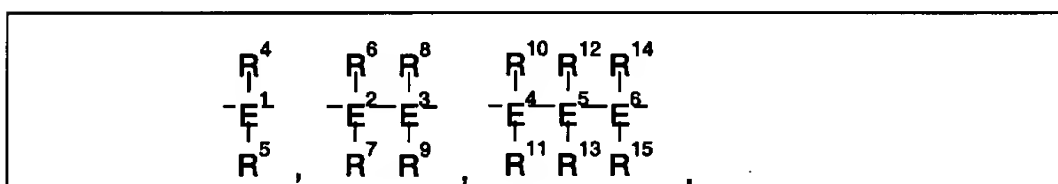
1. A process for the polymerization of olefins, which comprises carrying out the polymerization in the presence of catalysts comprising the following components:
 - (A) at least one complex of a transition metal with a tridentate macrocyclic ligand which bears at least one substituent having a donor function and
 - (B) optionally, one or more activator compounds.
2. A process as claimed in claim 1, wherein the component (A) is a compound of the formula I



where the variables have the following meanings:

M is a transition metal of groups 3 to 12 of the Periodic Table,

B¹-B³ are each a divalent radical selected from the group consisting of



where

E¹-E⁶ are silicon or carbon and not more than two of E⁴-E⁶ are silicon,

A¹-A³ are nitrogen or phosphorus,

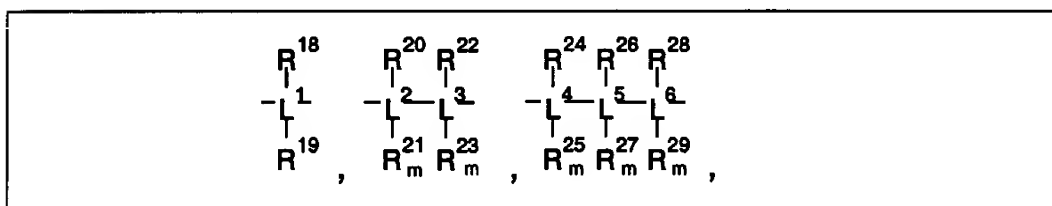
R¹-R¹⁵ are hydrogen, C₁-C₂₀-alkyl, 5- to 7-membered cycloalkyl which may in turn bear a C₆-C₁₀-aryl group as substituent, C₂-C₂₀-alkenyl, C₆-C₂₀-aryl,

alkylaryl having from 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part, SiR^{32}_3 or a radical of the formula $-\text{Z-D}$, where the organic radicals $\text{R}^1\text{-R}^{15}$ may be substituted by halogen(s) and any two geminal or vicinal radicals $\text{R}^1\text{-R}^{15}$ may also be joined to form a five- or six-membered ring, and at least one of the radicals $\text{R}^1\text{-R}^{15}$ is a radical $-\text{Z-D}$,

where

D is $\text{NR}^{16}\text{R}^{17}$, NR^{16} , OR^{16} , O, SR^{16} , S, $\text{PR}^{16}\text{R}^{17}$, SO_3R^{16} , OC(O)R^{16} , CO_2 , C(O)R^{16} , $\text{C(NR}^{16})\text{R}^{17}$, CN or a five- or six-membered heterocyclic ring system, where the radicals $\text{R}^{16}\text{-R}^{17}$ may also be joined to Z to form a five- or six-membered ring;

Z is a divalent radical selected from the group consisting of:



where

$\text{L}^1\text{-L}^6$ are silicon or carbon, not more than two of $\text{L}^4\text{-L}^6$ are silicon and $m=0$ if any two of the vicinal radicals R^{20} , R^{22} , R^{24} , R^{26} and R^{28} form an aromatic ring or a double bond is formed between two adjacent $\text{L}^2\text{-L}^6$, and otherwise $m=1$,

X are, independently of one another, fluorine, chlorine, bromine, iodine, hydrogen, $\text{C}_1\text{-C}_{10}$ -alkyl, $\text{C}_2\text{-C}_{10}$ -alkenyl, $\text{C}_6\text{-C}_{20}$ -aryl, alkylaryl having 1-10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part, $\text{NR}^{30}\text{R}^{31}$, OR^{30} , SR^{30} , SO_3R^{30} , OC(O)R^{30} , CN, SCN, =O, b-diketonate, BF_4^- , PF_6^- or bulky noncoordinating anions,

$\text{R}^{16}\text{-R}^{31}$ are hydrogen, $\text{C}_1\text{-C}_{20}$ -alkyl, 5- to 7-membered cycloalkyl which may in turn bear a $\text{C}_6\text{-C}_{10}$ -aryl group as substituent, $\text{C}_2\text{-C}_{20}$ -alkenyl, $\text{C}_6\text{-C}_{20}$ -aryl, alkylaryl having from 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part, SiR^{32}_3 , where the

organic radicals R^{16} - R^{31} may be substituted by halogen(s) and any two geminal or vicinal radicals R^{16} - R^{31} may also be joined to form a five- or six-membered ring,

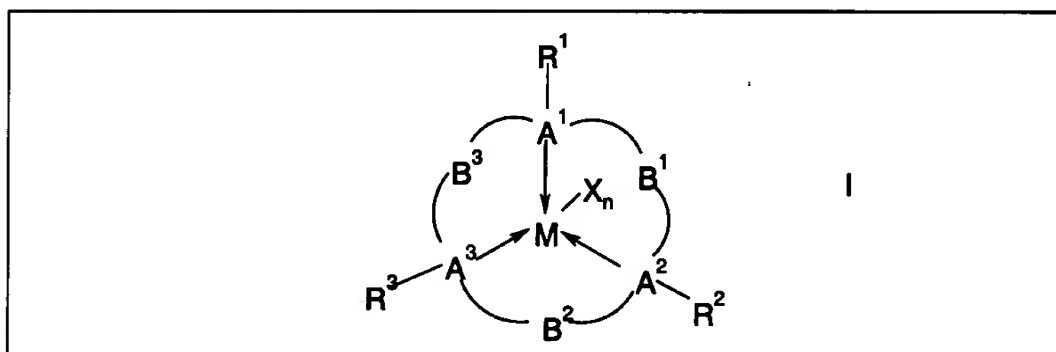
R^{32} are, independently of one another, hydrogen, C_1 - C_{20} -alkyl, 5- to 7-membered cycloalkyl which may in turn bear a C_6 - C_{10} -aryl group as substituent, C_2 - C_{20} -alkenyl, C_6 - C_{20} -aryl, alkylaryl having from 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part and any two geminal radicals R^{32} may also be joined to form a five- or six-membered ring,

n is a number from 1 to 4 which corresponds to the oxidation state of M or, if D is covalently bound to the metal center M , the oxidation state of M minus the number of groups D covalently bound to M , and, furthermore, the value of n is reduced by 1 for each X =oxygen.

3. A process as claimed in claim 2, wherein only R^1 is a radical -Z-D.
4. A process as claimed in claim 2, wherein B^1 , B^2 and B^3 are identical.
5. A process as claimed in claim 2, wherein D is oxygen, NR^{16} , $NR^{16}R^{17}$ or CN.
6. A process as claimed in claim 1, wherein the transition metal M comes from groups 3 to 8 of the Periodic Table.
7. A process as claimed in claim 1, wherein the transition metal M comes from group 6 of the Periodic Table.
8. A process as claimed in claim 1, wherein a compound selected from the group consisting of aluminoxane, dimethylanilinium tetrakis(pentafluorophenyl)borate, trityl tetrakis(pentafluorophenyl)borate and tris(pentafluorophenyl)borane is used as activator compound (B).
9. A process as claimed in claim 1, wherein at least one olefin selected from the group consisting of ethene, propene, 1-butene, 1-pentene, 1-hexene, 1-heptene or 1-octene is polymerized.
10. A process as claimed in claim 1, wherein the polymerization is carried out in suspension or in the gas phase.
11. A process as claimed in claim 1, wherein at least one metal complex (A) in the

presence of at least one catalyst (C) customary for the polymerization of olefins and, optionally, one or more activator compounds (B) is used.

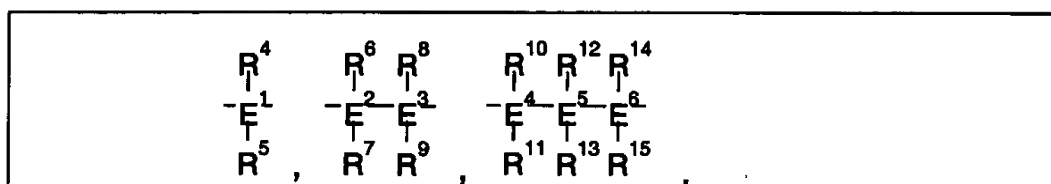
12. A catalyst system comprising the following components:
 - a) at least one transition metal complex (A) as defined in claim 1 and
 - b) at least one activator compound (B).
13. (canceled)
14. A process for the polymerization of olefins, which comprises carrying out the polymerization in the presence of catalysts comprising the following components:
 - (A) at least one complex of a transition metal with a tridentate macrocyclic ligand which bears at least one substituent having a donor function, wherein the macrocyclic ligand is covalently bonded to the transition metal and
 - (B) optionally, one or more activator compounds.
15. A process as claimed in claim 1, wherein the component (A) is a compound of the formula I



where the variables have the following meanings:

M is a transition metal of groups 3 to 12 of the Periodic Table,

B¹-B³ are each a divalent radical selected from the group consisting of



where

$\text{E}^1\text{-E}^6$ are silicon or carbon and not more than two of $\text{E}^4\text{-E}^6$ are silicon,

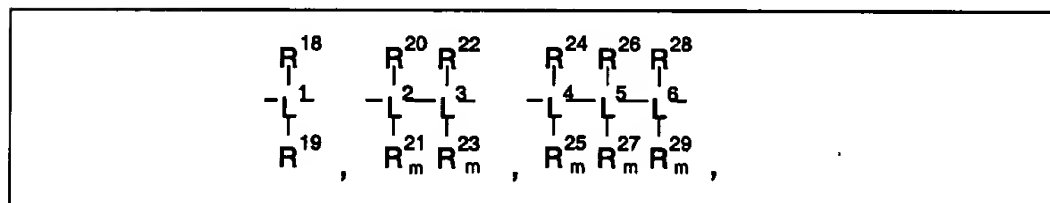
$\text{A}^1\text{-A}^3$ are nitrogen or phosphorus,

$\text{R}^1\text{-R}^{15}$ are hydrogen, $\text{C}_1\text{-C}_{20}$ -alkyl, 5- to 7-membered cycloalkyl which may in turn bear a $\text{C}_6\text{-C}_{10}$ -aryl group as substituent, $\text{C}_2\text{-C}_{20}$ -alkenyl, $\text{C}_6\text{-C}_{20}$ -aryl, alkylaryl having from 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part, SiR^{32}_3 or a radical of the formula $-\text{Z-D}$, where the organic radicals $\text{R}^1\text{-R}^{15}$ may be substituted by halogen(s) and any two geminal or vicinal radicals $\text{R}^1\text{-R}^{15}$ may also be joined to form a five- or six-membered ring, and at least one of the radicals $\text{R}^1\text{-R}^{15}$ is a radical $-\text{Z-D}$,

where

D is $\text{NR}^{16}\text{R}^{17}$, NR^{16} , OR^{16} , O, SR^{16} , S, $\text{PR}^{16}\text{R}^{17}$, SO_3R^{16} , OC(O)R^{16} , CO_2 , C(O)R^{16} , $\text{C(NR}^{16})\text{R}^{17}$, CN or a five- or six-membered heterocyclic ring system, where the radicals $\text{R}^{16}\text{-R}^{17}$ may also be joined to Z to form a five- or six-membered ring, and where D is covalently bonded to the transition metal M;

Z is a divalent radical selected from the group consisting of:



where

- L^1-L^6 are silicon or carbon, not more than two of L^4-L^6 are silicon and $m=0$ if any two of the vicinal radicals R^{20} , R^{22} , R^{24} , R^{26} and R^{28} form an aromatic ring or a double bond is formed between two adjacent L^2-L^6 , and otherwise $m=1$,
- X are, independently of one another, fluorine, chlorine, bromine, iodine, hydrogen, C_1-C_{10} -alkyl, C_2-C_{10} -alkenyl, C_6-C_{20} -aryl, alkylaryl having 1-10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part, $NR^{30}R^{31}$, OR^{30} , SR^{30} , SO_3R^{30} , $OC(O)R^{30}$, CN , SCN , $=O$, b-diketonate, BF_4^- , PF_6^- or bulky noncoordinating anions,
- $R^{16}-R^{31}$ are hydrogen, C_1-C_{20} -alkyl, 5- to 7-membered cycloalkyl which may in turn bear a C_6-C_{10} -aryl group as substituent, C_2-C_{20} -alkenyl, C_6-C_{20} -aryl, alkylaryl having from 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part, SiR^{32}_3 , where the organic radicals $R^{16}-R^{31}$ may be substituted by halogen(s) and any two geminal or vicinal radicals $R^{16}-R^{31}$ may also be joined to form a five- or six-membered ring,
- R^{32} are, independently of one another, hydrogen, C_1-C_{20} -alkyl, 5- to 7-membered cycloalkyl which may in turn bear a C_6-C_{10} -aryl group as substituent, C_2-C_{20} -alkenyl, C_6-C_{20} -aryl, alkylaryl having from 1 to 10 carbon atoms in the alkyl part and 6-20 carbon atoms in the aryl part and any two geminal radicals R^{32} may also be joined to form a five- or six-membered ring,
- n is a number from 1 to 4 which corresponds to the oxidation state of M or, if D is covalently bound to the metal center M , the oxidation state of M minus the number of groups D covalently bound to M , and, furthermore, the value of n is reduced by 1 for each X =oxygen.